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Characteristics of $PM_{2.5}$ Concentrations across Beijing during 2013–2015

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Abstract

High concentrations of particulate matter ($PM_{2.5}$) and frequent air pollution episodes in Beijing have attracted widespread attention. This paper utilizes data from the new air pollution network in China to examine the current spatial and temporal variability of $PM_{2.5}$ at 12 monitoring sites in Beijing over a recent 2-year period (April 2013) to March 2015). The long term (2-year) average concentration was 83 $\mu g \cdot m^{-3}$, well above Chinese and international standards. Across the region, annual average concentrations varied by 20 $\mu g \cdot m^{-3}$ (25% of the average level), with lower levels in suburban areas compared to periurban and urban areas, which had similar concentrations. The spatial variation in $PM_{2.5}$ concentrations was associated with several land use and economic variables, including the fraction of vegetated land and building construction activity, which together explained 71% of the spatial variation. Daily air quality was characterized as "polluted" (above 75 $\mu g \cdot m^{-3}$) on 36 to 47% of days, depending on site. There were 77 pollution episodes during the study period (defined as two or more consecutive days with Beijing-wide 24-hour average concentrations over 75 $\mu g \cdot m^{-3}$), and 2 to 5 episodes occurred each month, including summer months. The longest episode lasted 9 days and daily concentrations exceeded 450 $\mu g \cdot m^{-3}$. Daily $PM_{2.5}$ levels were autocorrelated ($r_{lag1} = 0.516$) and associated with many meteorological variables, including barometric pressure, relative humidity, hours of sunshine, surface and ambient temperature, precipitation and scavenging coefficient, and wind direction. Parsimonious models with meteorological and autoregressive terms explained over 60% of the variation in daily $PM_{2.5}$ levels. The first autoregressive term and hours of sunshine were the most important variables in these models, however, the latter variable is $PM_{2.5}$-dependent and thus not an explanatory variable. The present study can serve as a baseline to compare the improved air quality in Beijing expected in future years.

Keywords

Particulate matter; $PM_{2.5}$; spatial variation; temporal variation; Beijing; episode

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1 Introduction

Due to rapid economic growth, industrialization and urbanization, China has experienced severe air pollution problems, and Beijing, the capital, political and cultural center of China, is among the most polluted cities in the country. Air pollution has been recognized as a major concern in China (Ministry of Environmental Protection of the People's Republic, 2016), and both monitoring and emission controls have been ramped up in recent years. Concentrations of particulate matter with an aerodynamic diameter of 2.5 μm or smaller ($PM_{2.5}$) in Beijing have attracted global attention due to the high levels, as well as its associations with adverse effects on human (Adamkiewicz et al., 2015; Jakubiak-Lasocka et al., 2015; Zanobetti et al., 2014; Zhang et al., 2014a) and ecological health (Chen et al., 2014; Li et al., 2015b; Zhang et al., 2010). Many effective air pollution controls have been implemented, for example, during the 2008 Beijing Olympic Games, wide-ranging control measures were imposed that included shutting down pollution sources in the city and peripheral areas. While most of these measures have been discontinued, other emission controls have been implemented, such as fuel desulfurization at many large facilities. However, Beijing continues to experience a gray haze due to pollution, particularly in winter, and concentrations of SO_2, NO_2, PM_{10} and $PM_{2.5}$ remain high, e.g., 2014 averages were 21.8, 56.7, 115.9 and 85.9 $\mu g \cdot m^{-3}$, respectively (Beijing Municipal Environmental Protection Bureau, 2015). An especially lengthy and severe haze episode occurred in central and eastern China during January, 2013 during which Beijing, the most polluted area, experienced a 1-hr $PM_{2.5}$ concentration of 680 $\mu g \cdot m^{-3}$ (Wang et al., 2014a). This precipitated an orange haze alert, issued for the first time in Beijing (Ding and Liu, 2014). Unusual meteorological conditions were thought to be the main cause of this episode (Wang et al., 2014a; Zhang et al., 2014b). However, as discussed below, a similar episode occurred the following year.

The Chinese government has prioritized the improvement of air quality, and in February 2012 released the latest Ambient Air Quality Standards (GB 3095-2012) that established ambient $PM_{2.5}$ standards for the first time. These standards have two grades that apply to different areas. The Grade I $PM_{2.5}$ standard limits 24-hour daily and annual average concentrations to 35 and 15 $\mu g \cdot m^{-3}$, respectively, and applies to special regions such as national parks. The Grade II standard limits daily and annual averages to 75 and 35 $\mu g \cdot m^{-3}$, respectively, and applies to general areas. The Grade I standard is similar to the World Health Organization (WHO) Interim Target Three (IT-3), and the Grade II standard is equivalent to WHO Interim Target One (IT-1) (WHO, 2005). In China, air quality is considered "excellent" if 24-h average concentrations of $PM_{2.5}$ are below 35 $\mu g \cdot m^{-3}$, "favorable" for concentrations between 35 and 75 $\mu g \cdot m^{-3}$, and "polluted" for concentrations above 75 $\mu g \cdot m^{-3}$. When polluted, concentrations below 115 $\mu g \cdot m^{-3}$ are termed "lightly polluted", 115 to 150 $\mu g \cdot m^{-3}$ are "moderately polluted", 150 to 250 $\mu g \cdot m^{-3}$ are "heavily polluted", and concentrations above 250 $\mu g \cdot m^{-3}$ are "ultra-seriously polluted".

Under support by the Ministry of Environmental Protection, a nation-wide air quality monitoring network has been operating in China since January 2013. This modern network facilitates air quality investigations throughout the country, and helps identify effective preventive measures. The network features automatic online monitoring of six pollutants

($PM_{2.5}$, SO_2, NO_2, PM_{10}, CO, O_3), and was expanded in January 2014 from 550 sites in 74 cities to nearly 900 sites in 190 cities. Sites are classified as “urban,” typically at locations in cities designed to reflect urban air quality, and as “contrast” sites, which are located in upwind directions about 20 km from the city, thus reflecting “background” concentrations. The network includes three types of $PM_{2.5}$ monitoring: beta (β) attenuation using a dynamic heating system (DHS); light scattering; and tapered-element oscillating microbalance (TEOM) plus a filter dynamics measurement system (FDMS). $PM_{2.5}$ concentrations measured by these three approaches are similar to those from manual samplers (China National Environmental Monitoring Center, 2013).

This study analyzes the spatial and temporal characteristics of $PM_{2.5}$ concentrations in Beijing with the aims of characterizing pollution levels and air pollution episodes, and identifying factors that influence the spatial and temporal variation. We analyze daily data from 12 monitoring sites located across Beijing for a recent 2-year period (April 2013) to March 2015). Prior studies examining $PM_{2.5}$ in Beijing have been limited for several reasons, including the previously sparse monitoring network (Liu et al., 2015; Song et al., 2015; Yang et al., 2015a; Zhang et al., 2015b; Zhao et al., 2009), and the use of short or discontinuous monitoring periods (Duan et al., 2006; He et al., 2001; Yang et al., 2015b; Zheng et al., 2005).

2 Material and methods

2.1 Study sites

Beijing (39° 26' - 41° 03' N, 115° 25' −117° 30’ E) contains 14 districts and two counties, and occupies an area of 16,411 km^2 with an average altitude of 43.5 m above mean sea level. The population of Beijing was 21.148 and 21.516 million at the end of 2013 and 2014, respectively (Beijing Municipal Bureau of Statistics, 2013, 2014). Beijing consists of four functional zones (Figure 1): the ecological conservation development zone (Z1), which comprises three districts (Huairou, Mentougou, and Pinggu) and two counties (Miyun and Yanqing) and serves as an ecological barrier and water supply conservation area; the new urban development zone (Z2), which comprises five districts (Tongzhou, Shunyi, Daxing, Changping, and Fangshan) and is the base of the manufacturing industry and modern agriculture; the urban expansion zone (Z3), which contains four districts (Chaoyang, Haidian, Fengtai, and Shijingshan) and serves as the center of the modern economy and international communication; and the capital core zone (Z4), consisting of Dongcheng and Xicheng Districts, which is the political and cultural center of Beijing.

2.2 $PM_{2.5}$ data

Information from the 12 state-controlled ambient air quality monitoring sites across Beijing in the China National Air Quality Monitoring Network was utilized (Ministry of Environmental Protection of the People's Republic of China, 2016). These sites all use TEOMs plus FDMS. Each of the four functional zones contains at least one state-controlled monitor. The Dingling site (site B1) is defined as a contrast site, and the remaining 11 sites are urban sites (Figure 1). We calculated the Beijing-wide daily average concentration if at least 9 sites had valid daily concentrations. (Contrast site B1 was excluded from this average.) Using the Chinese air quality descriptors noted earlier, a pollution episode refers to a two day or longer period when the Beijing-wide daily 24-hour average concentration exceeded 75 $\mu g \cdot m^{-3}$. Hourly $PM_{2.5}$ data measured at the US Embassy in central Beijing (U.S. Department of State, 2016) also were obtained and compared to the state-operated network data. $PM_{2.5}$ concentrations at the US Embassy monitor and Beijing-wide levels had good agreement (e.g., r=0.895, n=689; Supplemental Figure 2), suggesting that the Beijing data are accurate and that concerns of "gaming" the numbers, expressed in earlier studies (Chen et al., 2012), are unwarranted.

2.3 Land use, economic, and meteorological data

Information regarding anthropogenic emissions of $PM_{2.5}$, land use, and economic data was obtained from the Beijing Statistical Information Net (Beijing Municipal Bureau of Statistics, 2016). Land use data included land area, cultivated land, garden plot, woodland, grassland, urban and industrial land, transport-related land, water area and the fractions of crop, grass, and forested land. Because Xicheng (D7) and Docheng Districts (D8) had incomplete data on "green" areas, an integrated parameter, called the "greening ratio" (GR), was constructed as the total of forest land, bush forest land, and forested areas near farms, roads, and rivers, divided by the total area. Economic data included local gross domestic product (GDP), which was divided into primary, secondary and tertiary industries, and output value and density for industries including construction industry and catering industry. Additional variables included energy consumption, represented by coal consumption, and permanent population and population density. In total, 23 variables were derived.

Meteorological data were obtained from the Chinese Meteorological Data Net (China Meteorological Administration, 2016). Variables included (or derived) were air temperature, surface temperature, wind speed, wind direction for the highest sustained (10 min) wind speed, wind direction for the highest instantaneous wind speed, daily precipitation, storm total precipitation, number of days in a rainfall event, atmospheric pressure, sunshine hours, and relative humidity. A daily scavenging ratio R_t was constructed as $R_t = 1 - \exp(-k\ P_t)$ where t = day, k = scavenging coefficient (nominally set to 2 mm^{-1}), and P_t = precipitation on day t. Several variables had minimum, maximum and average values (e.g., surface and ambient temperature and wind speed). Over 40 meteorological variables were considered. These data had no missing data.

2.4 Statistical analyses

After checking and cleaning, the data were summarized using descriptive statistics and trend plots. Initially, paired-sample *t* tests, Pearson correlation coefficients, simple regressions and scatterplots were used to investigate differences between urban sites and explore factors that might influence $PM_{2.5}$ levels. Factors potentially associated with the spatial variation in 2-year average $PM_{2.5}$ concentrations among the monitoring sites were investigated using multiple linear regression models (to test multiple factors simultaneously), the forward variable selection method, and the land-use and economic variables described earlier. We then evaluated relationships between meteorological factors and the temporal variation of daily $PM_{2.5}$ levels, again using correlations, scatterplots, and multiple regression models. The regression models tested a large number of variables, including lagged meteorological variables (e.g., P_t and R_t). Because daily concentrations were autocorrelated, a variety of time series models were evaluated, including autoregressive (AR) models with external inputs (i.e., meteorological variables) and Box-Jenkins time series models. Multiple imputation procedures (to replace missing $PM_{2.5}$ data) were used to generate the full dataset required for the AR models. No significant differences were noted between the Box-Jenkins and multiple regression models. Variable selection and model building were informed by the descriptive analyses and examination of residual plots with the goal of explaining the variation in $PM_{2.5}$ levels using parsimonious models and accepted mechanisms.

3 Results and discussion

3.1 Overview of $PM_{2.5}$ levels and standards attainment

$PM_{2.5}$ concentrations at the 12 sites are summarized in Table 1. Data completeness was very high, e.g., all stations except B7 had over 90% valid daily observations.

Daily $PM_{2.5}$ levels varied considerably, as shown in Figure 2. At individual sites, daily average concentrations ranged from 3 to 437 $\mu g \cdot m^{-3}$, and 2-year average concentrations from 69 to 89 $\mu g \cdot m^{-3}$. Mean concentrations exceeded medians, showing right-skewed distributions with several extremely high values and large standard deviations. The contrast site B1 had the lowest daily median and annual average concentrations, however, the maximum daily level at this site (377 $\mu g \cdot m^{-3}$) was similar to those at the other sites. Table 1 applies the Grade-II standards (same limit values as IT-1) with daily and annual $PM_{2.5}$ limits of 75 and 35 $\mu g \cdot m^{-3}$, respectively. The annual standard was exceeded at all sites, and the daily standard was exceeded from 34 (site B1) to 47% (site B7) of days.

Using the Chinese descriptors of pollution noted earlier, Beijing experienced heavy pollution on 8.3 to 11.3% of days ($PM_{2.5}$ levels from 150 to 250 $\mu g \cdot m^{-3}$), and ultra-serious pollution on 1.7 to 4.8% of days ($PM_{2.5}$ levels over 250 $\mu g \cdot m^{-3}$), depending on the site. Figure 3 shows classifications by site; as seen earlier, the contrast site (B1) had better air quality than elsewhere, e.g., 38.4% of days were classified as excellent, although 2.9% of days were considered ultra-seriously polluted. The many days when Grade-II standards were exceeded (Table 1), as well as the air quality classifications (Figure 3), indicate the poor air quality in Beijing during the study period.

$PM_{2.5}$ concentrations in Beijing are several or many times higher than levels in cities in Europe and the United States (Barmpadimos et al., 2012; Chow et al., 2006; Gehrig and Buchmann, 2003; Perrone et al., 2013; Querol et al., 2008; Russell et al., 2004), and two to three times higher than levels in Taiwan (Yu, 2010). However, $PM_{2.5}$ concentrations in Beijing have been decreasing in recent years. $PM_{2.5}$ levels exceeding 100 $\mu g \cdot m^{-3}$ were reported for 1999–2002 (Duan et al., 2006; He et al., 2001). Subsequently, Beijing implemented a number of air pollution control measures, e.g., between 2013 and 2014 emissions have been reduced by switching coal-fired boilers to cleaner fuels, shuttering other coal-fired plants, eliminating “yellow-label” and outdated vehicles, and switching coal residential heating to electricity (Beijing Municipal Government, 2015). A decline in $PM_{2.5}$ concentrations has been reported in several recent studies. Based on aerosol optical depth (AOD) in Beijing, a proxy for $PM_{2.5}$, an increasing trend was found from 2002 to 2006, followed by a strong decline from 2010 to 2012 (Zheng et al., 2015). Ground-based $PM_{2.5}$ data in Beijing show that annual mean $PM_{2.5}$ concentrations from 2005 to 2013 have declined by 31.8 and 13.3 $\mu g \cdot m^{-3}$ at urban and rural sites, respectively (Zhang et al., 2015b). More recently, annual average $PM_{2.5}$ concentrations across Beijing have fallen from 89.5 $\mu g \cdot m^{-3}$ (2013) to 85.9 and 80.6 $\mu g \cdot m^{-3}$ in 2014 and 2015, respectively (Beijing Municipal Government, 2015). Our Beijing-wide estimates are consistent with this downward trend, although a longer period should be considered to evaluate long-term trends.

3.2 Spatial variation of $PM_{2.5}$ and its influencing factors

Table 1 shows that $PM_{2.5}$ levels were lower at sites B1, B2, B3 and B4, which are located in the northern portion of the study region and 60 to 40 km from the capital core zone (Z4) that includes Dongcheng and Xicheng Districts (Figure 1). In contrast, the highest annual average concentration and the lowest fraction of days attaining standards occurred at site B7 in the Haidian district, which is in the western portion of the study region and about 12 km north of the core zone. Concentration differences were confirmed using paired-sample *t* tests and daily data. The contrast site B1 had levels 3 $\mu g \cdot m^{-3}$ (site B2) to 19 $\mu g \cdot m^{-3}$ (site B7) lower than the city sites ($p < 0.05$). In the large ecological conservation zone (Z1), where $PM_{2.5}$ was monitored at only one site (B2), average levels were 3 to 16 $\mu g \cdot m^{-3}$ lower than at the other sites. In the new urban development zone (Z2), $PM_{2.5}$ levels at two sites (B3 and B4) differed ($p < 0.05$), and both were significantly lower than concentrations in the periurban and urban districts. In the northern and western urban expansion zone (Z3), representing the periurban districts, concentrations differed between sites B5 and B6, B5 and B7, B6 and B8, and B7 and B8, but not between sites B5 and B8, and B6 and B7. In the capitol core zone (Z4), the center of Beijing, concentrations at the four sites (B9, B10, B11, B12) were similar (within 5 $\mu g \cdot m^{-3}$), and differences were not statistically significant between sites B9 and B10, and B10 and B11. Comparing functional zones, strong difference existed between sites Z1 and Z2, and Z3 and Z4, but not between Z3 and Z4. Ranking areas in terms of increasing $PM_{2.5}$ levels, Z1 < Z2 < Z3 ≈ Z4 ("≈" denotes no significant difference). Overall, $PM_{2.5}$ levels in suburban areas (Z2) were below levels in periurban and urban areas, while levels in periurban and urban areas were similar.

$PM_{2.5}$ concentrations have been associated with several types of land uses that serve as surrogates for the scale and intensity of anthropogenic activities (Wu et al., 2015). $PM_{2.5}$ concentrations at the Beijing area sites were negatively correlated to garden area ("green" space; $r = -0.89$, $p<0.001$), forest land area ($r = -0.81$, $p=0.001$), grass land ($r = -0.83$, $p=0.001$), and the greening ratio ($r=-0.72$, $p=0.002$). Land use may affect pollutant levels in many ways, e.g., vegetation (particularly trees) may capture some $PM_{2.5}$, and areas with a high fraction of vegetation may have fewer sources of emissions (e.g., industry and major roads). While such land use and economic factors may help explain some of the spatial variation in 2-year average concentrations across Beijing, the difference between the highest and lowest concentrations was only 20 $\mu g \cdot m^{-3}$, representing only 25% of the city-wide concentration of 83 $\mu g \cdot m^{-3}$.

Industry, traffic and building construction activities can be important $PM_{2.5}$ sources (Ianniello et al., 2011; Sun et al., 2004; Wang et al., 2013; Yu and Wang, 2010). Measures of district primary, secondary and tertiary industry output and GDP, as well as density measures of these variables (e.g., output per square kilometer) had low to moderate correlation with $PM_{2.5}$ levels, e.g., the correlation coefficients between $PM_{2.5}$ concentrations and district coal consumption was 0.24 (p=0.37), and 0.61 (p=0.01) for GDP. $PM_{2.5}$ levels were negatively correlated to tertiary industry output (r=0.58, p=0.02) and to lodging and catering output (r=0.56, p=0.03). Although the permanent population was not significantly correlated with $PM_{2.5}$ levels (r=0.42, p=0.11), population density was positively correlated with $PM_{2.5}$ (r=0.55, p=0.03). Fugitive dust, much of which is associated with construction activity, is another important $PM_{2.5}$ source (Hu et al., 2014). $PM_{2.5}$ concentrations had moderately high correlation to building industry output (measured in 10^8 Yuan, r = 0.63, p=0.01). While these results suggested the influence of population density, industry and green space, this analysis has several limitations: the metrics are only indirect and approximate surrogates of $PM_{2.5}$ emissions; emissions from tall stacks may affect concentrations at longer distances; the districts are not homogeneous; monitor concentrations are not necessarily representative of the entire district; and the sample size and diversity of sites are limited.

Overall, of the 23 factors examined, seven (greening ratio, GDP, tertiary industry output, building industry output, lodging and catering output, and population density) were significantly associated with the spatial variation of long-term $PM_{2.5}$ levels across Beijing. These were collapsed to two factors in a multiple linear regression model:

$$\mathrm{C_i}=88.89-0.176\ \mathrm{GR_i}+0.058\ \mathrm{BO_i} \qquad \mathrm{R}^2=0.71, \text{adjusted } \mathrm{R}^2=0.66 \quad (1)$$

where C_i = 2-year average $PM_{2.5}$ level in district I ($\mu g \cdot m^{-3}$), GR_i = greening ratio (fraction), and BO_i = building industry output (10^8 Yuan). In this model, which explained 71% of the spatial variation, a 10% increase in the district green ratio was associated with a decrease of $1.8 \pm 0.47\ \mu g \cdot m^{-3}$ in $PM_{2.5}$ concentration, and a 10^9 Yuan increase in building industry was associated with an increase of $0.58 \pm 0.20\ \mu g \cdot m^{-3}$.

3.3 Temporal variation, autocorrelation, and air pollution episodes

Daily $PM_{2.5}$ concentrations across the sites were highly correlated, e.g., correlation coefficients among site pairs ranged from 0.876 (sites B1 and B10) to 0.986 (B11 and B12). This temporal agreement occurs in large part to regional transport of $PM_{2.5}$ (DeGaetano and Doherty, 2004; Hu et al., 2014; Sun et al., 2006), which also is one of the main contributors to pollution episodes (Gao et al., 2013), especially in winter (Zhang et al., 2015a). During the 2008 Beijing Olympic Games, for example,Streets et al. (2007) estimated that 34% of $PM_{2.5}$ was due to sources outside of Beijing, in particular, the neighboring Hebei and Shandong Provinces and Tianjin Municipality. Wang et al. (2015) found nearly the same fraction (35.5% or 32.8 $\mu g \cdot m^{-3}$) for long-distance transport between 2005 and 2010, and also noted that the transport-related contribution increased by 1.2% annually over this period. Southern Hebei, Northern Henan and Southwest Shandong were identified as significant $PM_{2.5}$ contributors to pollution in Beijing (Zhang et al., 2015a). Wang et al. (2014b) estimated that during the heavy haze episode of January 2013, cross-city transport outside the Beijing-Tianjin-Hebei (BTH) region contributed 20 to 35% of $PM_{2.5}$, and transport among cities within the BTH region contributed 26% to 35% of $PM_{2.5}$, both compared to emissions within the BTH region. The sizable contribution from regional sources indicates the need to control both regional and local emissions sources to improve air quality in Beijing (Hu et al., 2014).

The daily $PM_{2.5}$ data have moderately high autocorrelation. For the Beijing-wide daily averages, lag 1- to 3-day autocorrelation coefficients were 0.516, 0.143, and −0.020, respectively, and partial autocorrelation coefficients were 0.516, −0.168, and −0.028. respectively. (Hourly data have higher autocorrelation, e.g., the lag 1, 2 and 3 partial autocorrelation coefficients at the US Embassy site were 0.741, 0.243 and 0.135, respectively.) A simple lag 1 autoregressive (AR1) model was the single best (highest R^2) one variable model for the daily Beijing-wide $PM_{2.5}$ levels:

$$\mathrm{C_t}=0.522\mathrm{C_{t-1}}+39.634 \qquad \mathrm{R}^2=0.27 \quad (2)$$

where C_t = $PM_{2.5}$ concentration on day t. A similar AR2 model slightly improved model fit:

$$\mathrm{C_t}=0.611\mathrm{C_{t-1}}-0.168\mathrm{C_{t-2}}+46.670 \qquad \mathrm{R}^2=0.30 \quad (3)$$

The AR structure, which indicates that today's pollution level depends in part on the previous days' levels, is an important explanatory factor in models predicting daily $PM_{2.5}$ levels, as shown later.

Figure 2, shown earlier, depicts trends of daily Beijing-wide concentrations and highlights periods with ultra-seriously polluted (>250 $\mu g \cdot m^{-3}$) and more favorable (<75 $\mu g \cdot m^{-3}$) conditions. Of the 704 days with sufficient data to determine the city-wide concentrations, 306 were considered classified as "polluted" using the Chinese description of greater than >75 $\mu g \cdot m^{-3}$. Figure 3 shows the classification at each monitoring sites. High concentrations occurred on several or many days every month (Figure 4). There were 2 to 5 air pollution episodes each month (at least two consecutive days when the Beijing average concentration exceeded 75 $\mu g \cdot m^{-3}$). $PM_{2.5}$ levels tended to be lowest in summer (particularly in August), and highest in winter, a pattern frequently observed elsewhere (Dimitriou and Kassomenos, 2014; Jhun et al., 2013; M.Marcazzan et al., 2001; Yang, 2002). Winters in Beijing are associated with conditions that prevailed in the "classical" air pollution episodes of the earthly and mid-20th century, namely, high emissions of particulate and gaseous pollutants from coal consumption for heating and industry, and frequent stagnation conditions due to subsidence inversions that impede pollutant dilution and transport (Hu et al., 2013; Shi et al., 2003; Zhao et al., 2009). In addition, Beijing's Spring Festival in late January and early February can increase $PM_{2.5}$ level due to extensive use of fireworks and firecrackers.

A total of 77 pollution episodes lasting a total of 267 days occurred during the 2-year study period. Episodes lasting 2, 3, 4, 5, 6, 7 and 9 days occurred 26, 21, 16, 5, 2, 6 and 1 times, respectively. The longest (9-day) episode occurred in summer (July 26 to August 3, 2014); several back-to-back episodes also occurred, including one discussed below. The long episodes showed a consistent pattern: a dramatic and rapid increase in $PM_{2.5}$ levels, which were sustained or even increased for a number of days, followed by a very rapid decline in $PM_{2.5}$ levels. This pattern was most apparent in winter. An especially severe case started on February 11, 2014 when "excellent" $PM_{2.5}$ levels rapidly degraded (within 24 hours) to "ultra-seriously" polluted conditions; after a 3-day hiatus, this 7-day episode was followed by a second 7-day episode that started on February 20, 2014. Figure 5 shows trends of pollutant and meteorological variables over this period. The lack of spatial variation across the monitoring sites (shown by the cat-and-whisker plots) during the episodes is notable, i.e., all monitors, including the background site B1, had very similar trends. (Supplemental Figure 2 shows trends during two other episodes which occurred in the summer.) In January 2013, just prior to the present study, a similar 7-day episode occurred with a maximum hourly concentration of 680 $\mu g \cdot m^{-3}$; the "explosive" and "sustained growth" of $PM_{2.5}$ in this episode was partly attributed to rapid transformation of gaseous pollutants to secondary aerosols (Wang et al., 2014a).

3.4 Meteorological influences on $PM_{2.5}$

In summer, $PM_{2.5}$ concentrations in Beijing tended to be lower, however, lengthy and serious pollution episodes still occurred. Summers in Beijing characteristically have high precipitation, high temperature, high relative humidity, and low wind speeds (Figure 4). With the exception of precipitation, these conditions can contribute to the rapid production and accumulation of secondary aerosols. $PM_{2.5}$ levels were slightly or moderately correlated to many meteorological variables. (Supplemental Figure 3 plots Beijing-wide daily concentrations versus daily measures of six meteorological variables.) For example, daily $PM_{2.5}$ levels tended to decrease with wind speed ($r = -0.27$, $n=701$, Supplemental Figure 3A), and increase with relative humidity ($r=0.455$, Supplemental Figure 3C). Similar patterns have been seen elsewhere (Hien et al., 2002; Li et al., 2015a). High wind speeds can favor plume spread and dilution, while high relative humidity can promote secondary PM formation. However, simple correlations do not account for interactions among meteorological variables (e.g., frontal storms may arrive from particular wind directions accompanied with high winds), the autocorrelation in pollutant levels discussed earlier, and the possible influence of very high pollutant levels on the weather variables themselves. After discussing simple correlations, we use multivariate and autoregressive models to help disentangle these effects.

Precipitation can affect $PM_{2.5}$ concentrations (Barmpadimos et al., 2012). In Beijing, most (70%) precipitation occurs during the summer, and over the study period, there were 110 days with precipitation, including 79 days with light showers (<10 mm), 21 days with moderate rain (10.0 - 24.9 mm), 8 days with heavy rain (25.0 - 49.9 mm), and two major rain storms (July 8, 2013 with 84.2 mm; and September 2, 2014 with 106 mm). Daily precipitation amounts had negligible correlation with daily Beijing-wide $PM_{2.5}$ levels ($r=0.053$; Supplemental Figure 3B), and $PM_{2.5}$ concentrations on wet days (any precipitation, 79.6 ± 47.1 $\mu g \cdot m^{-3}$, $n=106$) did not differ significantly from levels on dry days (83.8 ± 67.5 $\mu g \cdot m^{-3}$, $n=595$). Interestingly, rain occurred in the midst of several severe pollution episodes, most notably a 7-day episode starting June 24, 2013 when the daily Beijing-wide $PM_{2.5}$ average reached 292 $\mu g \cdot m^{-3}$ (this episode included two days with 17 and 6 mm of rain), and a following 9-day episode starting June 30, 2014 when the daily $PM_{2.5}$ concentration reached 194 μg·m^{-3} (one day with 49 mm of precipitation; Supplemental Figure 2A). Only in the latter episode did $PM_{2.5}$ levels fall substantially with precipitation. $PM_{2.5}$ concentrations were slightly more likely to decrease with larger precipitation amounts, e.g., for daily precipitation totals exceeding 10 mm, and for precipitation events lasting 3 or 4 days (Supplemental Figure 4). The effect of precipitation on $PM_{2.5}$ concentrations did not vary seasonally.

The Beijing data, as well as the literature, show that associations between precipitation and $PM_{2.5}$ levels are quite variable (Hien et al., 2002; Li et al., 2015a; Zhang et al., 2015b). Precipitation can affect PM by several mechanisms, e.g., wash-out and rain-out will reduce concentrations in the air column, and surface wetting will inhibit entrainment of surface dust from roads and fields (Li et al., 2015b). On the other hand, precipitation may increase concentrations as the associated humidity can promote the formation and growth of airborne particles (correlation between daily precipitation and relative humidity in Beijing was $r = 0.285$, $p=0.002$), and precipitation may be associated with weather patterns that limit dispersion of pollutants, such as low ceiling heights and low wind speeds, thus increasing concentrations from local (and possibly regional) emission sources. These effects may depend on season, site, aerosol size distribution and composition, precipitation intensity and duration, among other factors.

In Beijing, the daily average air temperature was weakly correlated to the daily Beijing-wide $PM_{2.5}$ concentration ($r=-0.107$, $p=0.001$, $n=701$; Supplemental Figure 3D); the ground surface temperature was more strongly correlated ($r=-0.128$, $p=0.001$). On colder days (surface temperatures below 5 C), temperature was positively correlated to $PM_{2.5}$ levels ($r = 0.118$, $p=0.098$, $n=197$); on warmer days (above 15 C), the correlation was negative ($r= -0.176$, $p=0.001$, $n=376$; Supplemental Figure 6). The increased $PM_{2.5}$ concentrations at low surface temperatures may reflect additional emissions due to space heating. However, $PM_{2.5}$ levels were found to increase in summer, especially June, due to biomass burning in Beijing and environs (Zhao et al., 2009). Temperature influences PM levels by several mechanisms: fuel use (and emissions) associated with heating increases at low temperatures; convection and dispersion of pollutants from especially local sources can be affected by temperatures; and formation rates of secondary aerosols increase with temperature. Given these competing factors, it is not surprisingly that the study (and the literature) show varying relationships between $PM_{2.5}$ concentrations and temperature, e.g., negative correlation at low temperatures and positive correlation at high temperatures were shown in a European study (Barmpadimos et al., 2012), negative correlations were found during summer in Shijiazhuang (Li et al., 2015a) and in the Sichuan Basin (Li et al., 2015b), and a positive correlation was found in winter and spring in Beijing (Zhao et al., 2014).

The daily number of hours with sunshine in Beijing (defined as the hours with over 125 W/m^2) and the daily Beijing-wide $PM_{2.5}$ concentrations were negatively correlated ($r= -0.472$, $p=0.000$, $n=701$; Supplemental Figure 3D), agreeing with earlier work in Beijing (Zhang et al., 2015b). Among all meteorological variables, hours of sunshine had the strongest correlation to $PM_{2.5}$. Days with extensive sunshine are typically associated with conditions favorable for pollutant dispersion, e.g., unstable conditions (Zhang et al., 2015b).

However, high $PM_{2.5}$ levels increase light extinction and reduce insolation intensity, which can decrease the number of hours classified as sunny, thus, this variable may not be independent of $PM_{2.5}$ levels. For this reason, we removed sunshine from the multivariate models explaining the daily variation of $PM_{2.5}$ (described below).

Barometric pressure did not show statistically significant correlations with $PM_{2.5}$ levels (r=0.035; Supplemental Figure 3F). Considering wind direction, Figure 6 shows both pollution and wind "roses" that use the daily Beijing-wide $PM_{2.5}$ levels and the wind direction occurring during the daily maximum sustained wind speed. Concentrations tended to be highest with winds from the east and south, although these wind directions were relatively uncommon. The wind direction analysis was limited given the available (24-hour) data; the use of resultant wind direction and hourly data would be more informative.

A day-of-week analysis showed somewhat higher levels on weekends, and a month-of-the-year analysis showed peaks in February and October (Supplemental Figure 7). However, these effects depended on season and year, and they disappeared in the multivariate models discussed next.

Multiple linear regression and autoregressive models used to examine the effect of multiple meteorological variables on daily Beijing-wide $PM_{2.5}$ levels employed forward step-wise variable selection, given the large number of variables available and of interest. These models preferentially selected the autoregressive (AR) terms and then the number of hours of sunshine as "independent" variables. Given that sunshine may depend on $PM_{2.5}$ levels and thus may not be independent, models without this variable were also evaluated. In addition, we isolated the contribution of the AR terms by fitting models without these terms. We considered both complete models that used all selected variables, as well as more parsimonious models that had 8 or fewer independent variables.

Table 2 shows the "parsimonious" (8-variable) models for three cases. The "full" model case, which considered all meteorological and AR variables selected, included daily sunshine, barometric pressure, average humidity, the surface temperature (both average and minimum), the precipitation scavenging ratio, and lag 1 and 2 day AR terms. All terms were statistically significant. This model had an adjusted R^2 = 0.606, and most predictions were near the 1:1 line (Figure 7A). The analogous complete model, which included 14 variables (adding the precipitation storm total, minimum ambient temperature, lag 1 precipitation scavenging ratio, and wind direction sectors 1, 5 and 10), had slightly higher fit (adjusted R^2 = 0.623; Supplemental Table 1). In contrast, the 8-variable model that excluded the two AR terms added the lag 1 scavenging ratio and the average wind speed. In this model, the adjusted R^2 fell to 0.471, and overall model fit markedly deteriorated (Figure 7B). This and the analogous complete models (Supplemental Table 1) show the importance of autocorrelation; the lagged precipitation added into this model also suggests its importance. The third case excluded the sunshine variable. This caused the lag 1 scavenging ratio and wind sector 1 (winds from the north) to enter the model; in this model, winds from the north are associated with a 41 μg m^{-3} concentration drop. Compared to the 8-variable full model, the adjusted R^2 slightly decreased to 0.572, and additional scatter and over-prediction were apparent (Figure 7C). However, this model (plus the complete model in Supplemental Table

1) highlights the key meteorological variables associated with $PM_{2.5}$ levels in Beijing: barometric pressure, relative humidity, temperature, precipitation scavenging, and wind direction. Interestingly, most models did not include wind speed or day-of-week effects. These multivariate models presented are not unique, and models using other combinations of variables can perform similarly. The models highlight some of the key variables affecting $PM_{2.5}$ levels while accounting for interactions between meteorological variables and autocorrelation. In addition, these statistical models can be used in forecasting and pollution alerting systems, providing an alternative to more complex emission and dispersion model-based approaches.

The selected variables and model fit can depend on the variable selection method, the site considered, the type of variables considered, the specification of the model, and the fitting method (although robust methods provided similar results). We considered only a few interactions in deriving variables and fitting models, and variable transformations were mostly limited to wind speed, direction, surface temperature and precipitation variables. These statistical models could benefit from more direct (rather than surrogate) measures of $PM_{2.5}$ transport, emissions, and dispersion potential, as well as additional and site-specific meteorological data.

4 Conclusion

The frequent air pollution episodes and poor air quality in Beijing in recent years warrant attention and analysis. This study examined spatial and temporal variability of $PM_{2.5}$ in the Beijing area, emphasizing daily data collected over a recent 2-year period. $PM_{2.5}$ concentrations across the Beijing area averaged 83 $\mu g \cdot m^{-3}$, over twice the Chinese annual average standard. Long-term $PM_{2.5}$ concentrations varied by 20 $\mu g\ m^{-3}$ across the study area, and levels were lowest in the ecological conservation zone and the suburban area; differences between periurban and urban areas were small. Most of the spatial variation in $PM_{2.5}$ levels could be explained by green space and building industry activity, however, the spatial variation accounted for less than a quarter of annual average $PM_{2.5}$ levels. Daily data showed that pollution episodes classified as "serious" and "ultra-serious" occurred every month in Beijing. Episodes were characterized by a rapid increase in $PM_{2.5}$ concentrations, which were sustained for as long as a week and sometimes longer, followed by a rapid decrease in $PM_{2.5}$ levels. $PM_{2.5}$ concentrations exhibited seasonal patterns (generally higher in winter, lower in summer), and strong associations with several meteorological variables, including barometric pressure, relative humidity, temperature, precipitation using a scavenging ratio), and wind direction. Multivariate models with autoregressive terms explained over 60% of the variance in daily $PM_{2.5}$ levels. The single most important variable in predicting daily $PM_{2.5}$ concentrations was the previous day's $PM_{2.5}$ level. While the daily number of hours of sunshine was strongly associated with $PM_{2.5}$ levels, very high $PM_{2.5}$ levels may reduce the number of sunshine hours, thus, this variable may not be independent of the $PM_{2.5}$ concentration. Finally, data from the state-operated monitors had close agreement to measurements at the US Embassy.

While $PM_{2.5}$ concentrations across Beijing remain high, substantial improvements have occurred in recent years due to the policies and measures implemented to control air pollution. In September 2013, the Chinese government released the “Plan of Action for Preventing and Controlling of Atmospheric Pollution” (State Council of China, 2013) that implemented ten control measures, e.g., reducing coal energy consumption by using cleaner alternatives such as gas and water power. The Hebei–Beijing–Tianjin region was required to reduce $PM_{2.5}$ concentrations to 60 $\mu g \cdot m^{-3}$ by 2017. To achieve this goal, the Beijing municipal government issued a 5-year Clean Air Action Plan (2013–2017) that included policies to cut vehicle and industrial emissions. This Plan calls for reducing annual coal consumption by 13 million tons and reducing coal consumption to 10 million tons by 2017, promoting the use of clean energy in public vehicles (buses, taxis, and postal trucks), and reducing production at polluting facilities in the iron, steel, cement, chemical and petrochemical industries (Beijing Municipal Government, 2015). The present study can serve as a baseline to compare the improved air quality in Beijing that is expected in future years.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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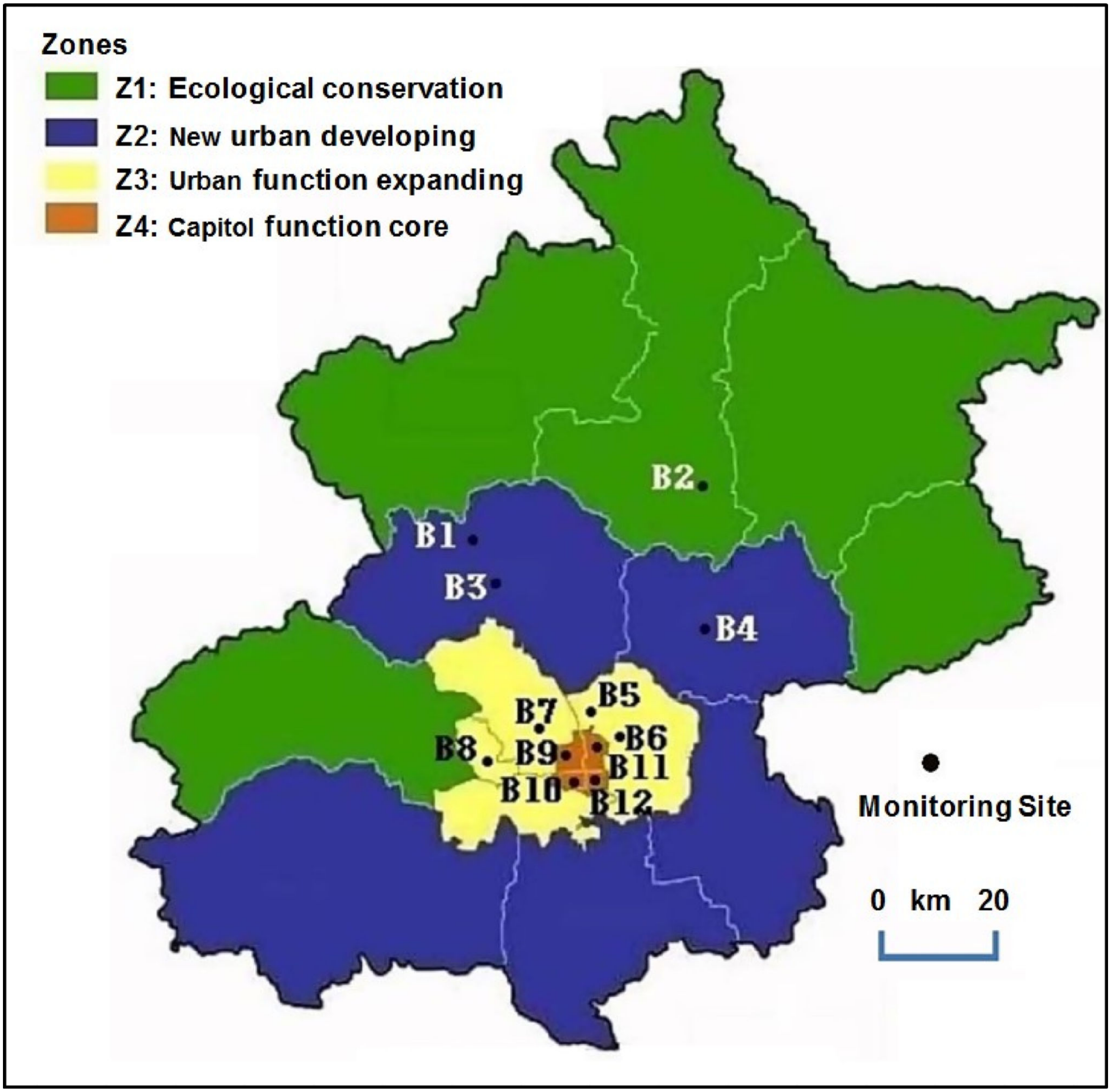

Figure 1.
Locations of the 12 air quality monitoring sites and 4 designed zones in Beijing.

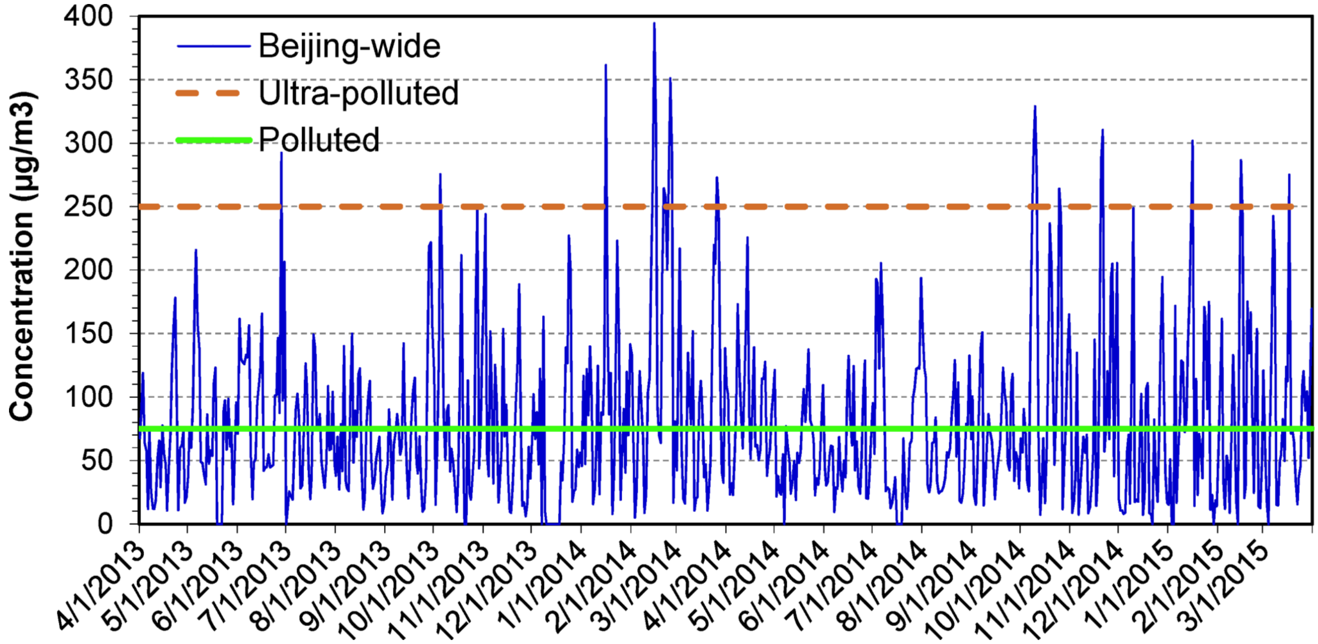

Figure 2.

Trends of daily Beijing-wide $PM_{2.5}$ concentrations from April 2013 through March 2014. Lines show daily limits of 75 and 250 μg m^{-3} corresponding to classifications of "polluted" and "ultra-polluted" days.

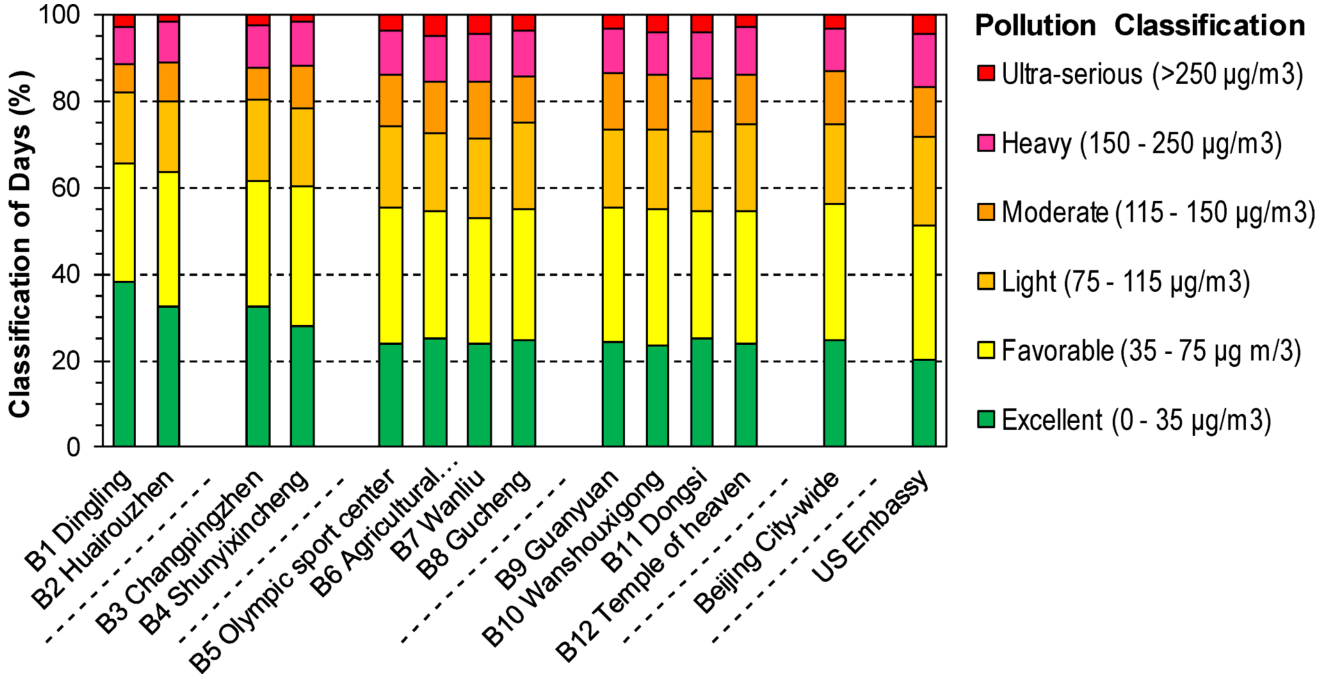

Figure 3.

Classification of days by $PM_{2.5}$ air quality levels. State operated sites grouped by zone.

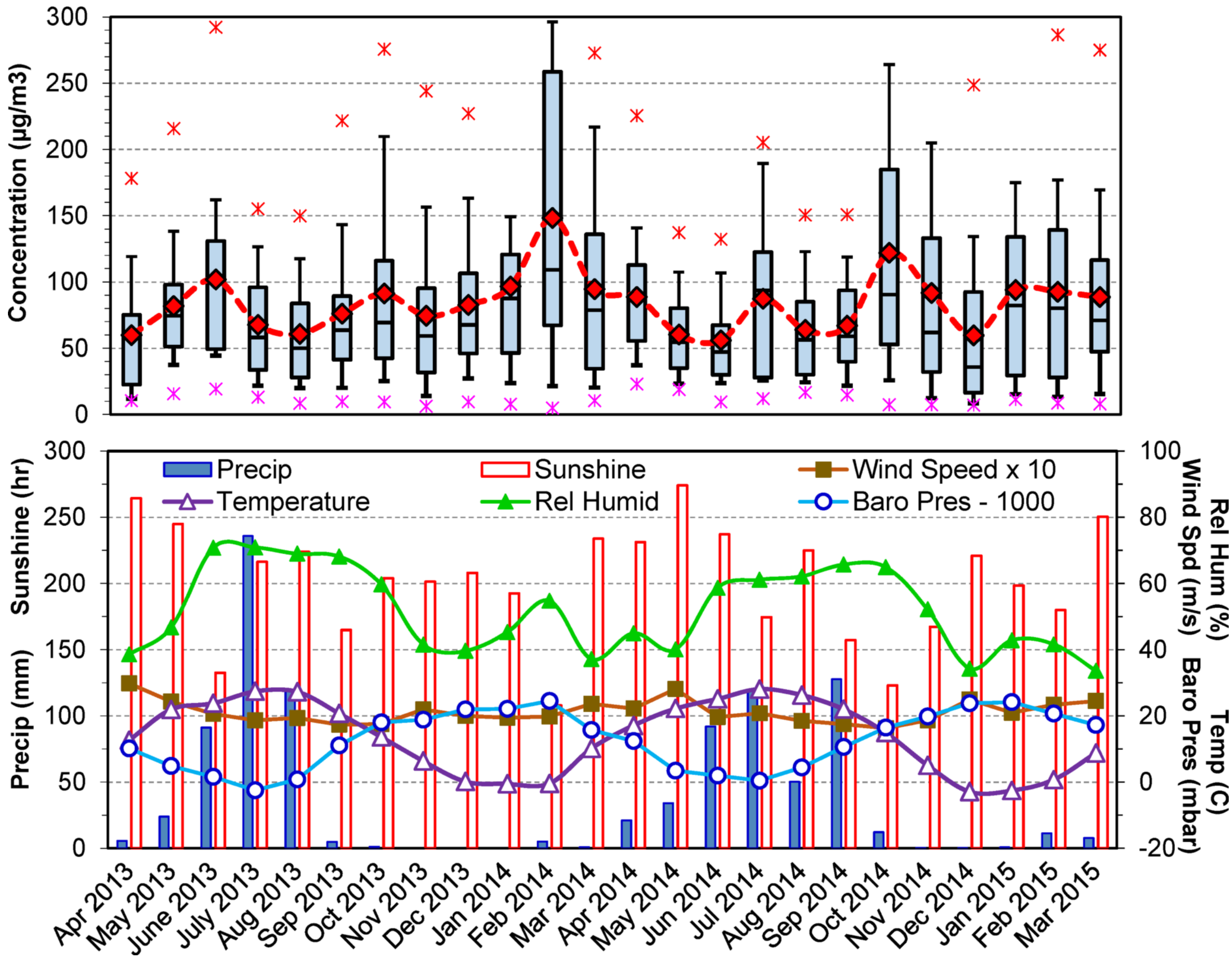

Figure 4.

Top: Monthly variation of Beijing-wide $PM_{2.5}$ levels from April 2013 through March 2015. Box and whisker plots show monthly 90th, 75th, 50th, 25th and 10th percentiles; maximum and minimum indicated as points; line (diamond symbol) shows monthly average. Bottom: Monthly variation of meteorological variables in Beijing, including monthly average temperature, relative humidity, wind speed, and barometric pressure; and monthly total precipitation and hours of sunshine.

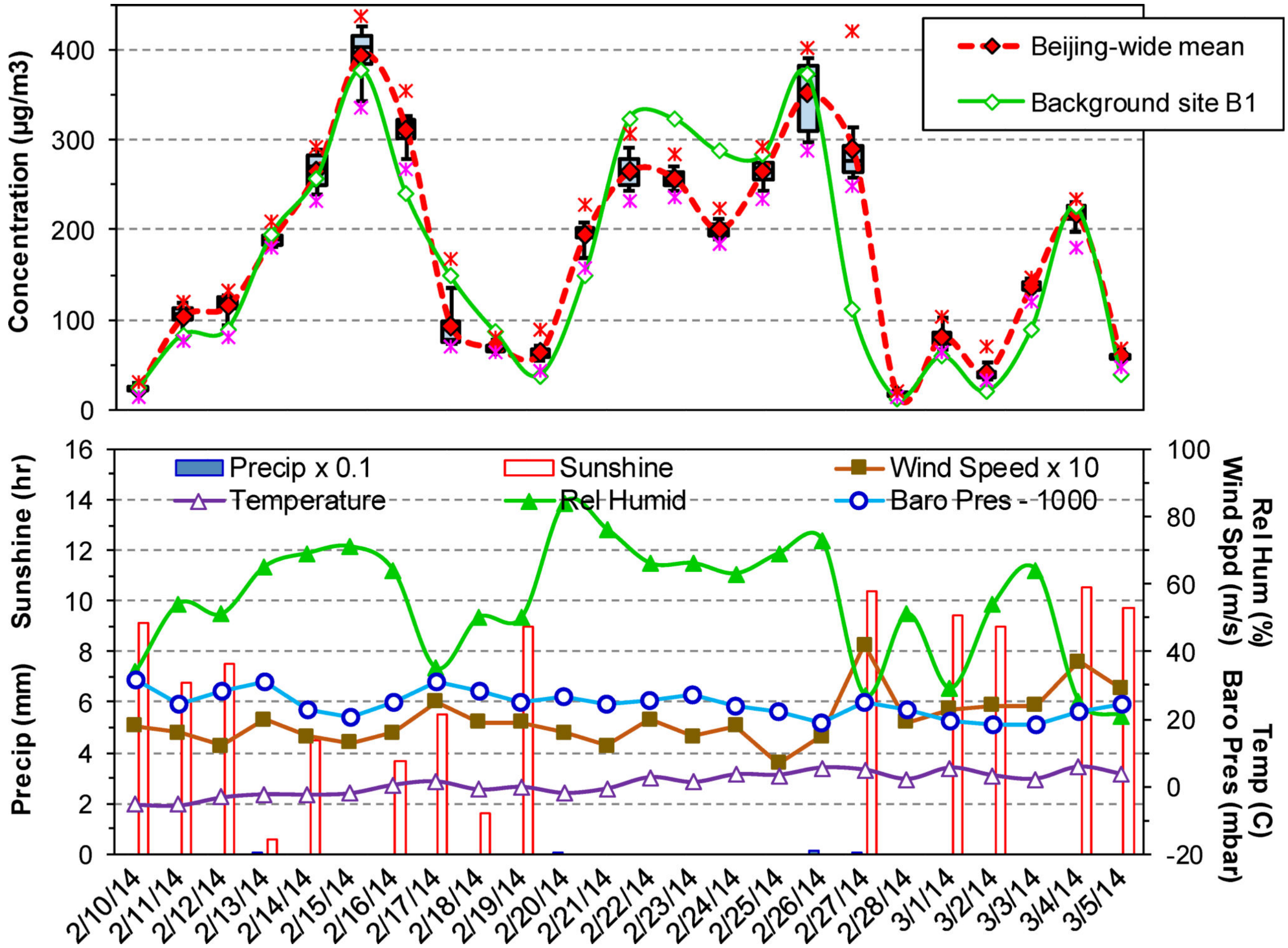

Figure 5.

Trends of $PM_{2.5}$ and meteorological variables over two pollution episodes in February 2014. Top: Daily Beijing-wide concentrations, including maximum, minimum, average, 10th, 25th, 50th, 75th, 90th percentiles; and daily concentration at background site B1. Bottom: Daily average of temperature, relative humidity, wind speed and barometric pressure, and daily sum of sunshine hours and precipitation.

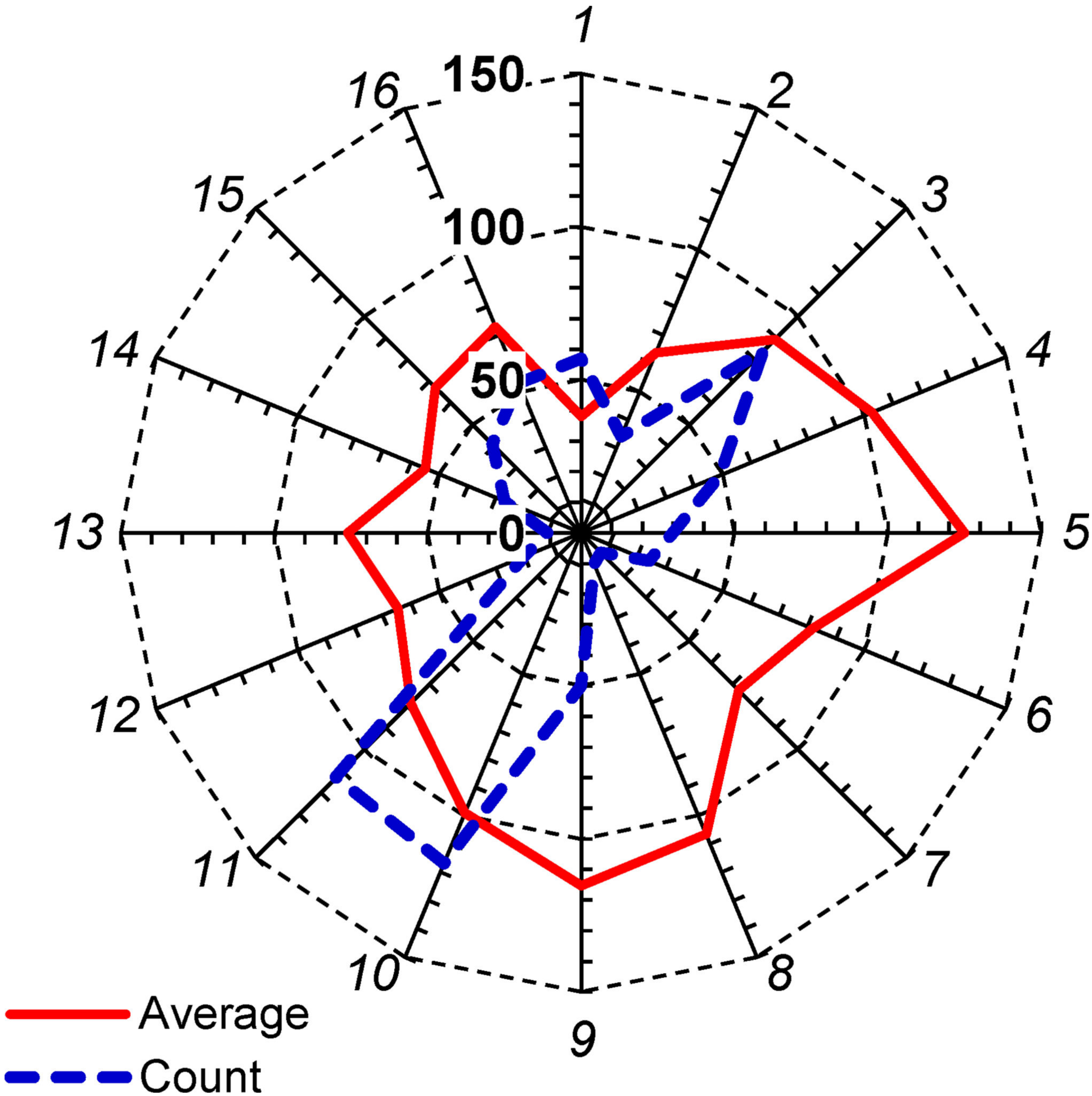

Figure 6.
Beijing-wide $PM_{2.5}$ concentrations as a function of wind direction. “Average” is average concentration within that sector; “Count” is number of days within study period within that sector. Sector 1 is north. Directions based on maximum sustained (10 min) daily wind speed.

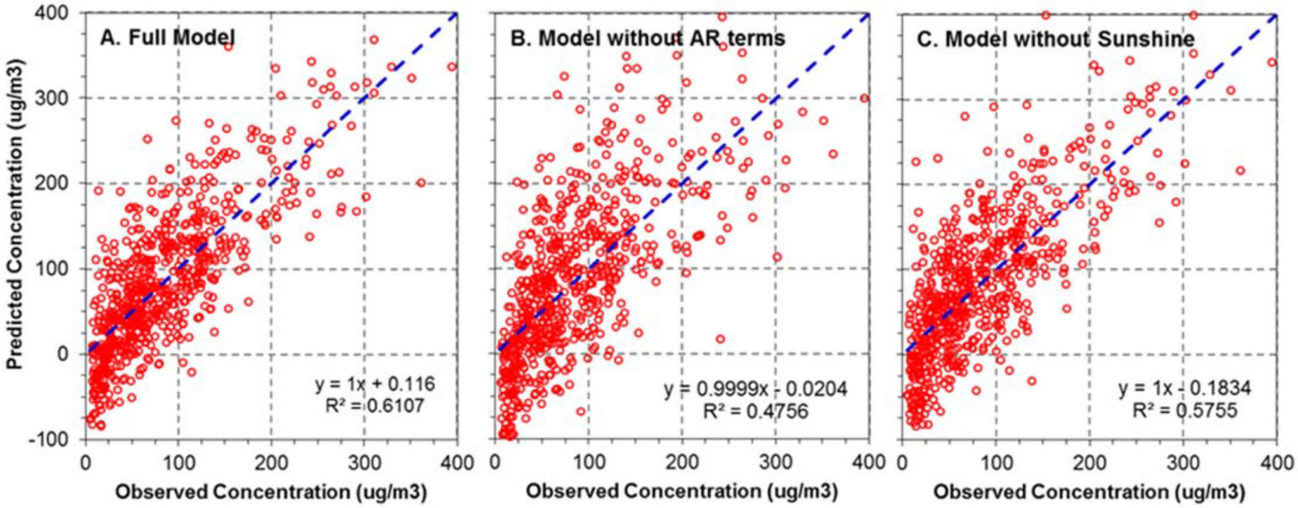

Figure 7.

Predicted $PM_{2.5}$ concentrations using "best" 8 parameter models, with and without autoregressive terms and number of hours of sunshine variables.

Table 1

Summary of daily $PM_{2.5}$ concentrations at the 13 monitoring sites. Beijing city-wide is average of 11 Beijing state-operated sites (excludes site B1).

	Zone 1		Zone 2		Zone 3				Zone 4					
Statistic	**B1 Dingling**	**B2 Huairouzhen**	**B3 Changpingzhen**	**B4 Shunyixincheng**	**B5 Olympic sport center**	**B6 Agricultural exhibition**	**B7 Wanliu**	**B8 Gucheng**	**B9 Guanyuan**	**B10 Wanshouxigong**	**B11 Dongsi**	**B12 Temple of heaven**	**Beijing City-wide**	**US Embassy**
Statistics														
Mean	69	72	75	77	85	87	89	85	85	86	87	85	83	94
St. Dev.	65	61	64	60	68	72	71	66	67	69	70	66	65	73
Minimum	4	3	3	4	5	6	3	5	4	4	3	5	5	7
Median	49	53	56	61	66	67	70	67	68	68	68	66	66	72
Maximum	377	421	364	407	401	421	417	395	396	437	425	407	394	450
NOBs	721	692	707	693	679	706	646	691	669	687	713	702	702	717
Days attaining standards, targets, or goals (%)														
Grade I (35 μg m^{-3})	39	34	34	28	24	25	25	26	25	24	26	24	25	20
Grade II/ IT-1 (75 μg m^{-3})	66	64	62	60	56	55	53	56	56	55	55	55	56	51
IT-2 (25 μg m^{-3})	31	23	24	20	17	18	15	16	18	16	19	17	17	12
IT^{-3} (15 μg m^{-3})	17	12	11	9	9	9	8	7	8	7	10	8	7	4
Air Quality Goal (10 μg m^{-3})	9	6	6	5	3	4	3	3	3	3	5	4	3	1

NOBs is number of valid observations. Grade I and Grade II standards refer to the China Ambient Air Quality Standards (GB 3095-2012). IT-1, IT-2, IT-3 and Air Quality Goal refer to the WHO interim targets and air quality guideline (AQG).

Table 2

Three regression models predicting Beijing-wide daily PM2.5 concentrations. “Full model” considers all variables. “Model without AR terms” excludes autoregressive terms. “Model without sunshine variable” excludes variable providing daily number of hours of sunshine. All models scaled to fit data.

	Full Model			Model without AR terms			Model without Sunshine		
	Coef.	Std.Err.	p-value	Coef.	Std.Err.	p-value	Coef.	Std.Err.	p-value
CONSTANT	1285.88	203.13	0.000	1222.93	228.42	0.000	1393.66	200.65	0.000
BP	−2.22	0.53	0.000	−2.80	0.77	0.000	−2.55	0.56	0.000
RH	3.26	0.21	0.000	5.18	0.34	0.000	3.67	0.19	0.000
TEMPS_AVE	7.12	0.99	0.000	9.09	1.48	0.000			
TEMPS_MIN	−10.89	1.03	0.000	−14.43	1.56	0.000	−4.21	0.49	0.000
PRECIP_SCAV	−89.74	9.90	0.000	−124.43	14.76	0.000	−87.96	10.73	0.000
PM_L1	0.75	0.05	0.000				0.83	0.05	0.000
PM_L2	−0.23	0.05	0.000				−0.20	0.05	0.000
SUNSHINE	−8.88	0.98	0.000	−13.81	1.45	0.000			
PRECIP_SCAV_L1				−38.73	13.48	0.004	−37.59	10.07	0.000
WD1							−40.87	11.62	0.000
WS				15.49	5.29	0.004			
N / R2 / Adj R2	676	0.611	0.606	701	0.477	0.471	676	0.576	0.570